Directional off-Normal Photon Streaming from Hybrid Plasmon-Emitter Coupled Metasurfaces

Yinhui Kan, Fei Ding, Changying Zhao,* and Sergey I. Bozhevolnyi*

ABSTRACT: Efficiently funnelling photon streams from quantum emitters (QEs) in controllable directions is essential for interfacing the photon emission from nanoscale volumes with complex optical configurations exploited in quantum information and communication systems. We propose hybrid plasmon-QE coupled metasurfaces featuring circular nanoridges with displaced centers to funnel the photon emission along a given off-normal direction. The design principle relies on phase matching QE-excited circularly diverging surface plasmons to a well-collimated off-normal propagating photon stream by using a modified bullseye antenna with circular nanoridges having appropriately displaced centers. We demonstrate with the simulations that the hybrid plasmon-QE coupled metasurfaces enable highly directional off-normal photon emission with collection efficiencies exceeding 96%. Using nanodiamonds with multiple nitrogen vacancy centers, we experimentally demonstrate the directional photon streaming with different, up to 17.3°, off-normal angles and efficiencies reaching 90% for solid angles of <0.1 sr.

KEYWORDS: quantum emitter, plasmonics, unidirectional emission, collection efficiency, fluorescence

Efficient collection of photons radiated by quantum emitters (QEs) is crucial for many quantum technologies, including quantum entanglement, quantum computation, and sensing. Typical unmodified QEs, such as molecules, quantum dots, and defects in diamonds, feature electric-dipolar emission patterns with poor directionality, thus, resulting in relatively low collection efficiencies that would hinder QEs being directly used in practical applications. To realize the QE directional emission, one would resort to either geometrical (far-field) or near-field beam-shaping approaches. The geometrical approaches typically involve redirecting and reshaping of the far-field QE emission by its focusing, reflecting, and refracting with mirrors and lenses, thereby unavoidably reducing the collection efficiency and requiring the use of bulky optical components. Alternatively, the near-field coupling approaches rely on the nonradiative QE interaction with surrounding nanostructures, thus, enabling a direct and efficient control of the far-field QE emission with highly compact configurations.

In recent years, QE coupling to nanoantennas/nanocavities has emerged as one of the most rapidly growing research topics in nano-optics. Nanopatch antennas have been demonstrated to enhance the spontaneous emission rate (the so-called Purcell effect), while also modifying and, to some extent, improving the QE emission directivity. Yagi-Uda antennas have been found useful for funneling the QE emission into a specific direction by placing a QE near the feed element. However, the resulting emission is not well collimated, featuring a relatively large angular dispersion and thus resulting in a relatively low collection efficiency. QE-coupled bullseye gratings formed by periodic concentric rings were introduced for improving the collection efficiency. Different kinds of bullseye gratings have been proposed, including bullseye apertures in metal films, metal bullseye structures, and hybrid plasmonic bullseye antennas. Among them, the hybrid plasmonic bullseye antennas, composed of dielectric nanoridges on metal substrates with dielectric spacers, appear most attractive from the viewpoint of combining high quantum yield and collection efficiency. At the same time, the QE emission is concentrated around the normal to the sample surface due to the bullseye symmetry, limiting the design flexibility with respect to the control over the QE emission direction, which is in turn highly desired in quantum optics and nanophotonics.

Here, we propose and demonstrate hybrid plasmon-QE coupled metasurfaces based on a modified bullseye antenna with circular nanoridges having appropriately displaced centers, a configuration that facilitates the phase matching of QE-excited circularly diverging surface plasmon polaritons (SPPs) and a well-collimated off-normal propagating photon stream by using a modified bullseye antenna with circular nanoridges having appropriately displaced centers. We demonstrate with the simulations that the hybrid plasmon-QE coupled metasurfaces enable highly directional off-normal photon emission with collection efficiencies exceeding 96%. Using nanodiamonds with multiple nitrogen vacancy centers, we experimentally demonstrate the directional photon streaming with different, up to 17.3°, off-normal angles and efficiencies reaching 90% for solid angles of <0.1 sr.
stream. First, the design principle is introduced with qualitative considerations and supported with numerical simulations. In the subsequent experiments, we assemble the hybrid plasmon-QE coupled metasurfaces by using nanodiamonds (NDs) containing nitrogen vacancy centers (NV centers) as QEs, which are surrounded by precisely positioned dielectric bullseye-displaced antennas. The experimental results are in excellent agreement with numerical simulations, demonstrating highly directional off-normal photon emission with high collection efficiencies.

The schematic and operation principle of the plasmon–QE coupled system are shown in Figure 1. A QE, situated in the center of the inner ring, is adjacent to the metal substrate to spontaneously decay through the excitation of SPPs propagating along the dielectric–metal interface. SPPs are scattered and subsequently converted into a stream of free-space light by the bullseye-like Doppler nanoridges having the ridge height with different periods on two sides, resulting in unidirectional off-axis emission. Figure 1b shows the cross-section of the structure, which is composed of the hydrogen silsesquioxane (HSQ) bullseye-like Doppler gratings on top of a silica covered silver film. The 20 nm thin silica spacer layer used here can not only mitigate the quenching of QEs but also protect silver from being oxidized. The SPPs excitation would dominate the decay channel along the dielectric

\[ \text{characteristic time-domain} \times = 2s \text{pp}, \]  

and the SPPs are scattered and subsequently converted into photons unidirectionally in the y-direction with \( \theta \). Here, \( G_{x,0} = 2\pi/P_{x,0} \) and \( k_{x,\theta} = \sin \theta k_{x,0} \), where \( k_{x,0} = 2\pi/\lambda \) is the wavevector of the out-coupling light in free-space and \( \theta \) is the angle of the emission. We would like to note that the above rigorous conditions for the realization of identical sloping angles for the outcoupled radiation can only approximately be satisfied (for small displacements and sloping angles) in our simplified design based on the usage of displaced circular ridges. In the following, we limit the considered displacements by 200 nm, a value that we estimated to be close to the crossover between the single- and double-beam outcoupling regimes for our configuration (see Supporting Information, section 1).

We perform the three-dimensional (3D) simulation for the proposed systems using the finite-difference time-domain (FDTD) method. Figure 2a shows the schematic of the top view of the structure, which consists of 8 nanoridges with width \( w = 180 \text{ nm} \). When there is no displacement of the nanoridges in x direction, the structure would degenerate into normal bullseye rings with the default period \( P = 550 \text{ nm} \). The displacement in x direction is defined as \( \Delta = (P + P_{1})/2 \), where \( P_{2} = P + \Delta \), \( P_{1} = P - \Delta \), and \( P = 550 \text{ nm} \), viewed as a QE, is vertically orientated on the top of the substrate with a 50 nm distance and situates in the center of the inner nanoridge with the radius of \( r_{m} = 520 \text{ nm} \). The refractive index of HSQ is set as 1.41. The permittivities of SiO\(_2\) and Ag are taken from ref 41. Let us first check the near-field electric field distribution of the coupled systems in two different cross sections. Figure 2b presents the electric field distribution in the y–z plane (position indicated by the dashed line A in Figure 2a). It shows that a portion of the SPPs are scattered out of the plane when meet the inner gratings, while the rest part continues propagating along the surface until being converted into photons by outer gratings or decayed by the metal loss. It demonstrates that the symmetrically distributed rings in y direction convert the SPPs into outcoming photons with a symmetric shape. While for asymmetric Doppler gratings along the x direction with displacement \( \Delta = 0.2 \mu \text{m} \), the SPPs are scattered and converted into photons unidirectionally in the x–z plane.

Figure 1. (a) Schematic of unidirectional and off-axis emission from a hybrid plasmonic-QE coupled system. (b) The cross-section view of the system. The gratings with different periods on two sides can unidirectionally outcouple the counterpropagating SPPs into oblique angles. (c) Schematic of the symmetric unidirectional outcoupling in the wave vector domain, resulting in two outcoupled waves propagating under different angles. Red and blue peaks represent, respectively, the right- and left- propagating SPP waves. Arrows indicate the coupling direction. (d) Schematic of the synchronized asymmetric outcoupling that results in single wave propagating under an oblique angle.

\[ \begin{align*}
G_{1} & = k_{\text{pp}} + k_{x,\theta} \\
G_{2} & = k_{\text{pp}} - k_{x,\theta}
\end{align*} \]
In the far-field pattern shown in Figure 2d, it is clearer that the outcoming photons in free space assembles a collimated bright spot in an off-axis direction, with the divergence angle of only $\approx 3^\circ$, as determined at the full-width-at-half-maximum (fwhm) of intensity distribution. The sloping unidirectional angle of the spot with respect to the center is $16.2^\circ$. The white circle indicates the collective angle of the objective with NA = 0.9.

We would like to note that, although the horizontally excited QE can also realize the similar direction emission (the results are not presented here), the decay rate of the QE is several times lower than the vertical regime shown here.

To experimentally demonstrate the unidirectional and off-axis emission, we fabricated the proposed systems with $\sim 100$ nm-diameter NDs, containing on average $\sim 400$ NV centers per ND (Adamas Nanotechnologies). These NDs can easily be located with the dark-field images, which makes it possible to precisely fabricate nanoridges around them (see Supporting Information). The substrate is fabricated by thermal evaporation of 3 nm Ti and 200 nm Ag on a Si wafer. Then, a 20 nm thick SiO$_2$ layer is deposited through RF-sputtering. NDs are spin coated on the surface after the gold aligning markers being fabricated with standard electron-beam lithography (EBL). The relative position of a selected ND with respect to prefabricated gold aligning markers is determined using the corresponding dark-field microscopy image. The HSQ circular nanoridges with different position displacement $\Delta = 0, 0.05, 0.1, 0.15$, and 0.2 $\mu$m are subsequently fabricated around the selected NDs with the second EBL (see Supporting Information for fabrication details). As shown in Figure 3a–e, the selected NDs are situated in the center of the nanoridges, validating the accuracy in positioning. The NDs containing many NV centers would have many dipole momentums with random directions. To excite the vertically oriented dipole, we use a tightly focused radially polarized 10 $\mu$W pump cw-laser beam at the wavelength of 532 nm which produces a strong longitudinal electric field component at the focal plane. In particular, this optical pump configuration ensures that the excited NV centers (in the selected NDs) have sufficiently large projections of their radiative dipole transitions in the direction perpendicular to the surface, making thereby the experimental conditions similar to those used in our simulations (Figure 2). Upon excitation, the NDs decay to the SPPs on the dielectric–metal interface and then converted into the free-space emission by HSQ nanoridges (Figure 3f–j). The emission patterns with different structures are then taken from Fourier plane, as shown in Figure 3f–j. The white line indicates the collection boundary of the objective NA = 0.9. Figure 3a,f shows a specific case that the structure is in azimuthal symmetry, where

Figure 2. (a) Top view of the proposed coupled systems, consisting of 8 nanoridges with a vertical QE in the center. (b) The electric field distribution in the $y$–$z$ plane. The cross section is cut along the dashed line A in (a). (c) The electric field distribution in the $x$–$z$ plane. The cross-section is cut along the dashed line B in (a). (d) The simulation results of the far-field emission pattern. The white circle indicates the collective angle of the objective with NA = 0.9.

Figure 3. (a–e) SEM images of the fabricated hybrid plasmonic–QE coupled systems with different ridge displacements $\Delta = 0, 0.05, 0.1, 0.15$, and 0.2 $\mu$m in $x$ direction, respectively. The NDs are situated in the center of the inner rings with the HSQ nanoridges being fabricated around. (f–j) Corresponding Fourier plane images of the different samples. The white line indicates images are taken with an objective of NA = 0.9.
a doughnut-shape emission pattern is achieved in the far field. When the displacement is increased, the emission patterns could maintain concentrated spots, which is attributed to the azimuthal distribution of the rings. Moreover, the spot gradually shifts to aside with different unidirectional angles, demonstrating the tunability of the unidirectional emission. For the configuration with ridge displacement of $\Delta = 0.2 \mu m$ in Figure 3e, the experimental measured angle of the collimated spot is $17.3^\circ$, and the emission pattern coincides well with the simulation result shown in Figure 2d.

Figure 4a shows the spectra of the fabricated samples with different displacements. All of them have two small peaks around 575 and 637 nm, which are fingerprints for the neutral (NV$^0$) and negative charge state (NV$^-$$^-$). The fluorescence of the NDs has a large photon distribution around 670 nm. For better correspondence with the simulation wavelength, we used a 676 ± 15 nm band-passing filter to block light at other wavelengths, as well as the pump laser beam. Figure 4b presents the relationship between the ridge displacement and the outcoupling angle of the unidirectional emission. The experimental results for different $\Delta$ are in excellent agreement with the corresponding numerical simulations in the whole range of displacements (0–200 nm). The design outcoupling angles are consistent with both numerically simulated and experimentally measured values only for small displacements ($\leq 100$ nm), while progressively overestimating the outcoupling angles for larger displacements. As such, the discrepancy should be expected for large ($>100$ nm) displacements due to approximations involved in the simplified design approach (based on displaced circular ridges) that results in two phase-matched outcoupling directions corresponding to the two different periods, $P_1$ and $P_2$ (see Supporting Information, section 1). The fact, that the deviation seems to better follow the smallest (of two) outcoupling angle, may be ascribed to stronger SPP scattering in the forward direction$^{9,40}$ that would enhance the contribution from the large period ($P_2$) grating, but other important factors might be involved (see Supporting Information, section 1). The collection efficiency as a function of the circular ridge displacement is also calculated. It shows that with different displacements the collection efficiencies keep around 0.965, which is promising for efficiently extracting photons out from nanoscale plasmonic–QEs coupled system. Therefore, the proposed coupled systems can act as a functional interconnect between the QEs and free-space complicated optical systems. Based on the design, in the future, it is possible to achieve more active and flexible manipulation of the direction of QE emission by introducing phase-change materials$^{44,45}$ electrical gating$^{46}$ and modulating surrounding environment$^{47}$ into the coupled system.

In summary, we have proposed hybrid plasmonic–QEs coupled systems consisting of bullseye-like Doppler nanoridges with NDs situated in the center of the inner rings. We experimentally demonstrated that the proposed systems have the capability of directing the emission from NVs to a unidirectional and off-axis collimated beam in the far field. It shows that the unidirectional angle can be as large as $17.3^\circ$ with the circular ridge displacement of $\Delta = 0.2 \mu m$. The collection efficiency can maintain a high value about 0.965 with a collective objective of NA = 0.9. The off-axis angle is relevant to the position shift, demonstrating the ability of the proposed systems to actively guide the direction of the emission from QEs. This work paves the venue to flexibly connect the quantum emission with complex optical systems.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsphotonics.0c00196.

Qualitative analysis of our design approach; Numerical simulation to estimate the nanoridge period; Detailed sample fabrication description; Experimental setup used for characterization. Section S1: Numerical simulation for determining the wavelengths of SPPs and the default period of gratings. Section S2: Detailed description of the fabrication of the samples. Section S3: Schematic of the experimental setup (PDF)

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Notes
The authors declare no competing financial interest.

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Supporting Information

Directional off-normal photon streaming from hybrid plasmon-emitter coupled metasurfaces

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1. Qualitative analysis of our design approach

The conventional bullseye ridge configuration, which is used for outcoupling surface plasmon-polariton (SPP) waves excited by a quantum emitter (QE) in the direction perpendicular to the surface, consists of concentric circular ridges with the period, $P$, equal to the SPP wavelength. If one would like to outcouple the SPP waves in the off-normal direction in the $x$-$z$-plane (see Fig. 1 in the main text), the ridge periods along the $x$-axis on two sides of the origin (where the QE is located) should be modified accordingly, becoming thereby different:

\[
G_1 = k_{spp} + k_{x,\theta},
\]
\[
G_2 = k_{spp} - k_{x,\theta}.
\]

Here, $G_{1,2} = 2\pi/P_{1,2}$ and $k_{x,\theta} = \sin \theta k_0$, where $k_0 = 2\pi/\lambda$ is the wavevector of the out-coupling light in free-space and $\theta$ is the angle of the emission.

The above (rigorous) phase-matching conditions can readily be rewritten in the following form:

\[
\frac{P - P_1}{P_1 P} = \frac{P_2 - P}{P P_2} = \frac{\sin \theta}{\lambda}.
\]

Considering these conditions [Eq. (S3)] from the viewpoint of using our simplified design approach based on displaced circular ridges, it becomes transparent that these phase-matching conditions can only approximately be satisfied with our approach. In general, the problem of accurately satisfying the phase-matching conditions for off-axis beaming of surface waves is rather complicated. In our simplistic approach with $P - P_1 = P_2 - P = \Delta$, two different ridge gratings located on two sides of the origin, containing the QE source, would outcouple the SPP waves along different sloping angles:

\[
\sin \theta_{1,2} = \frac{\lambda \Delta}{P(P + \Delta)}.
\]
The arithmetic (mean) average of the above relations

\[
\sin \theta = \frac{\lambda^2}{p^2 - \Delta^2}
\]  

(S5)

can be used to evaluate the design outcoupling angle for a configuration of displaced circular ridges with given parameters. It can further be shown that the associated angle differences are small for small displacements that imply small sloping angles: \(\sin \theta_1 - \sin \theta \sim \sin \theta - \sin \thetaz \sim \frac{\Delta}{P}\) (here, we left out higher order terms).

In the practical realization, the number of circular ridges, \(N\), is limited, a circumstance that results in diffraction (angular) divergence of the outcoupled waves: \(\delta \theta \approx \frac{\lambda}{NP}\). This divergence should be compared with the sloping angle differences considered above in order to qualitatively estimate the critical displacement marking the crossover between the single- and double-beam outcoupling regimes for our configuration. We would like to emphasize that the real situation is much more complicated with many other important factors, such as stronger SPP scattering in the forward direction \(^{54}\) that would enhance the contribution from the large period \((P_2)\) grating, influence of the \(z\)-component of SPP and scattered waves, progressively increasing for larger outcoupling angles, and nonuniform amplitude distributions of the scattered waves due to radial SPP divergence, SPP absorption and SPP scattering out by ridges. Additionally, the filling ridge factor becomes spatially inhomogeneous for displaced ridges influencing the SPP wave vector magnitude (see Supporting Section 2) and, thereby, the phase matching condition. Considering our configuration \((P = 550 \text{ nm}, \lambda = 670 \text{ nm}, N = 8)\), it is seen that the differences can be neglected for the displacements < 100 nm corresponding to the sloping angles < 10° (Fig. S1). For the purpose of exploring the limits of our approach and bearing in mind the aforementioned complications we decided to study displacements of up to 200 nm (Fig. S1).
Figure S1. The dependences (solid lines) of outcoupling angles for two sides of the displaced circular ridges calculated using Eq. (S4) for different displacements $\Delta$ along with the average outcoupling angle given by Eq. (S5). Dashed lines indicate the angular spread of outcoupling angles due to the diffraction divergence of outcoupled waves.

2. Numerical Simulation

Numerical simulations in this work are performed by commercial FDTD software package (FDTD Solutions, Lumerical Solutions). An electric dipole, viewed as an QE, is vertical to the substrate surface situated 50 nm above the top film. The wavelength in the simulation is $\lambda_0 = 670$ nm, which is consistent with the peak emission of the negative charge state of the NV centers in NDs\textsuperscript{s1}. The permittivities of Ag and SiO\textsubscript{2} are taken from the handbook of optical constants of solids\textsuperscript{s5}, while the refractive index of HSQ is set as 1.41\textsuperscript{s6}. First, we conduct the 2D FDTD simulation to estimate the period of the bullseye gratings, which are used for scattering the QE-excited SPP waves to free space emission normal to the surface. In Fig. S2, we present the wavelength $\lambda_{spp}$ of SPPs and effective
indexes $N_{eff}$ for both the scenarios without or with HSQ layer on the top of substrate. The results show that for Air-SiO$_2$-Ag regime and HSQ-SiO$_2$-Ag regime the SPPs effective indexes $N_{eff1} = 1.101$ and $N_{eff2} = 1.446$, respectively. According to the effective medium theory, the effective index $N_{eff}^G$ of the dielectric grating can be weighted by the grating fill factor $\chi$ as $N_{eff}^G = (1 - \chi) N_{eff1} + \chi N_{eff2}$. For $\chi = 0.33$, we have $N_{eff}^G = 1.215$. Correspondingly, to scatter SPPs into free space emission the period of the grating should be set as $\Lambda = \frac{\lambda_0}{N_{eff}^G} \approx 550$ nm. To verify these parameters, we conduct the 3D FDTD simulation that the electric dipole situated in the center and surrounded by 8 circularly distributed HSQ nanoridges with $\Lambda = 550$ nm, $\chi = 0.33$. Figure S2(c) is the far-field intensity distribution of out-coupling emission, which demonstrates that under this design the SPPs can couple into free-space unidirectional beam with a doughnut pattern.

![Figure S2](image)

**Figure S2.** The wavelength and effective indexes of SPPs propagating along the interface between SiO$_2$ and Ag, (a) without HSQ layer (i.e. Air-SiO$_2$-Ag regime), (b) with HSQ layer (i.e. HSQ-SiO$_2$-Ag regime). The thickness of Ag substrate is 200 nm, followed by a 20 nm SiO$_2$ layer. The SPPs are excited by an electric dipole situated 50 nm above the SiO$_2$ layer surface and then propagate along the dielectric-metal interface. The dash line rectangles in the insets indicate the displayed areas in the simulation domain for presenting the distribution profiles of the electric field component $E_x$. (c) Far-field intensity distribution of out-coupling emission for 8 circularly distributed nanoridges with $\Lambda = 550$ nm, $\chi = 0.33$. 

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3. Sample fabrication

The substrates consist of three layers: 3 nm Ti and 200 nm Ag, topped by RF-sputtering of 20 nm SiO$_2$ on a Si wafer, which are fabricated by thermal evaporation at deposition rates of 0.1 Å/s, 1 Å/s, and 1.5 Å/s and $5 \times 10^{-6}$ mbar chamber pressure, respectively. The Ti film here acts as an adhesion layer. PMMA A2 is subsequently spincoated at 1580 rpm (45 s) on the substrates and prebaked at 180°C for 2 min before patterning align markers by a 30-kV electron beam lithography system (JEOL-6490). After development (1:3 MIBK-to-IPA for 35s followed by 1min rinse in IPA), 35 nm Au layers are deposited on the samples by ohmic evaporation at 1 Å/s and $5 \times 10^{-6}$ mbar chamber pressure. Lift-off is performed in acetone for 6 h to reveal the alignment markers. 100 nm NDs containing ~400 NV-centers (Adamas technology) are spincoated on the samples. The relative positions of the NDs in the coordinate frame of the alignment marks are determined by a dark-field microscope image (Fig. S3). A coordinates system is initially defined by selecting the lower left marker as the point of origin. Selecting the lower right marker, defines the x-axis and physical dimensions, by scaling pixel length to the known distance between selected markers. A well-defined $xy$-coordinate is confirmed by plotting the estimated position of the top markers. The ND coordinates is finally obtained by fitting two Gaussians to the diffraction-limited spot of the ND, as show in Figs. S3(g) and S3(h). Figure. S3(i) shows the precise position of the ND with accuracy to nanometer. This high accuracy is important in this work because the positions of the NDs in the inner ring is crucial to the coupling between SPPs and gratings. Then, an HSQ layer was subsequently spincoated at 1000 rpm (60 s) and prebaked at 160°C for 2 min. The position- shift bullseye gratings are patterned around the NDs by electron beam lithography by aligning to the makers, followed by development using 25% TMAH (Tetramethylammonium hydroxide) for 4 min to reveal the structures.
Figure S3. Alignment procedure for determining the position of a ND. (a) Starting point is dark-field image in grayscale, showing four alignment marks M1-M4 and the target ND. (b) A coordinate system is defined by setting the center of M1 to origin, indicated by red spot. (c) A Horizontal coordinate axis and scaling is defined by selecting the center of M2. (d), (e) A control of the defined coordinate system is done, by plotting expected positions of M3 and M4. (g), (h) The coordinate position of the ND is determined fitting two gaussians fitting (red) to the diffraction-limited spot of the ND (blue). (i) The final estimated position of the ND indicated by the green spot with the accuracy of nanometer.

Figure S4. Process for the fabrication of the samples.

4. Experimental setup

Figure S5 is the schematic of the optical set-up that is used for characterizing NDs-nanoridges coupled systems. The lamp is used to preliminarily find samples in the microscopy. The pump laser
is a linearly polarized 532 nm continuous wave (Crystal laser). A half-wave plate (λ/2) in combination with a Glan-Taylor polarizing beam splitter (PBS) allow for power control and lock the axis of linear polarization. A liquid crystal display (ARCoptix RPC; ARCoptix) converts the linear polarized light to a radially polarized beam which is used to drive the NDs. The beam is focused onto the sample using a x100 0.9 NA objective (MPLFLN x100; Olympus). Fluorescence collected by same objective is filtered from the laser light, by a set of dichroic mirrors (DM) (FF535-SDi01/FF552-Di02; Semrock) and a long pass filter (LPF) (FELH0550; Thorlabs). Recording the fluorescence photon rate with an avalanche photo diode (APD) (τ – SPAD, Pico quant), while scanning the sample, using a piezo-stage, allowed for locating NV-centers by the recording of fluorescence maps. The spectrum of the NV-center was measured with a spectrometer equipped with a CCD camera (Ultra 888 USB3 –BV, Andor). A flip mirror projects the Fourier plane onto a CCD camera (Orcad4LT; Hamamatsu) to obtain the Fourier plane images.

**References**


